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LABORATORY EVALUATION AND APPLICATION OF
MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED
CONDITIONS FOR PLANETARY ATMOSPHERES

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Submitted by

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from spacecraft radio occultation experiments and earth-based radio astronomical observations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpretation of available opacity data. For example, results obtained for the microwave opacity from gaseous H_2SO_4 under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from H_2SO_4 much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). Subsequent measurements made by Steffes and Jenkins (1987), showed that the microwave opacity of gaseous ammonia (NH_3) under simulated Jovian conditions did indeed agree with theoretical predictions to within experimental accuracy at wavelengths longward of 1.3 cm. Work performed by Joiner et al. (1989) during the fourth and fifth years of Grant NAGW-533 and continuing into this past grant year (February 1, 1989 through October 31, 1989) has shown that laboratory measurements of the millimeter-wave opacity of ammonia between 7.5 mm and 9.3 mm and also at the 3.2 mm wavelength require a different lineshape to be used in the theoretical prediction for millimeter-wave ammonia opacity than had been previously used. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and

pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

A key activity for the past grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Semiannual Status Report #11 for Grant NAGW-533 (February 1, 1989 through April 30, 1989), initial laboratory measurements of the millimeter-wave opacity of gaseous ammonia (NH_3) in a hydrogen/helium (H_2/He) atmosphere, under simulated conditions for the outer planets were completed in 1988. These measurements were conducted at frequencies from 32 to 40 GHz (wavelengths from 7.5 to 9.3 mm). A complete description of this millimeter-wave spectrometer is given in previous Annual Status Reports for Grant NAGW-533, and in Joiner et al. (1989).

Since larger variations from theoretically-derived opacity values were expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we have conducted laboratory measurements at wavelengths near 3.2 mm (94 GHz), where a large number of observations of the emission from the outer planets have been

made. A description of this new system was presented in Semiannual Status Report #11 (February 1, 1989 through April 30, 1989). A better knowledge of the millimeter-wave absorption properties of NH_3 is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as H_2S (see Bezard et al., 1983). This knowledge will be of considerable importance for millimeter-wave instruments proposed for future missions, such as the MSAR (microwave and spectrometer and radiometer) proposed for the Cassini mission.

In some cases, new observations or experiments have been suggested by the results of our laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid (H_2SO_4) under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and early Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere (Steffes, 1985). Further laboratory measurements also suggested that a substantial variation in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2.2 cm wavelength. Since no observations of the Venus emission at this wavelength had ever been published, we conducted observations of Venus using the 140-foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation were substantial in that they not only placed

limits on the abundance and spatial distribution of gaseous H_2SO_4 and SO_2 , but they also suggested some limits to long term temporal variations for the abundance of these two gases.

Recently, we completed calibration and interpretive studies on the data from these observations and a paper entitled "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P.G. Steffes, M.J. Klein, and J.M. Jenkins, has been accepted by the journal Icarus. One important issue which was discussed in this paper is the discovery that the microwave absorptivity for gaseous H_2SO_4 which was measured by Steffes (1985 and 1986) appears to differ from a theoretical spectrum newly computed from over 11,000 lines by Janssen (personal communication) by a scale factor. That scale factor suggests that the theoretically-derived "dissociation factor" for gaseous H_2SO_4 (i.e., the percentage of H_2SO_4 which breaks down to form SO_3 and H_2O) may have been underestimated. This results in an underestimation of the absorption from gaseous H_2SO_4 . Therefore, an experiment has been conducted to correctly evaluate the "dissociation factor" and thus allow unambiguous calibration of laboratory data for H_2SO_4 opacity (see Section III). This will be critical for the proper interpretation of a wide range of opacity data.

Another important tool for evaluating potential spatial and temporal variations in abundance and distribution of gaseous H_2SO_4 is the reduction and analysis of recently obtained Pioneer-Venus radio occultation measurements. The 13 cm microwave absorptivity profiles, which can be obtained from the radio occultation data, are closely related to the abundance profiles for gaseous H_2SO_4 . Starting in June 1988, we began the reduction of the 1986-87 Pioneer-Venus radio

occultation measurements (working at JPL with support from the Pioneer-Venus Guest Investigator Program) in order to obtain the needed 13 cm microwave absorptivity profiles. Yet another important source of information is the increasing number of high-resolution millimeter-wavelength Venus emission measurements which have been recently conducted. Correlative studies of these measurements with radio occultation measurements and our longer wavelength emission measurements (Steffes et al., 1989) should provide the necessary data for characterizing temporal and spatial variations in the abundance of gaseous H_2SO_4 and SO_2 , and for modeling its role in the subcloud atmosphere. However, unambiguous results will require that we have dependable knowledge of the equilibrium between gaseous H_2SO_4 , SO_3 , and H_2O , both so as to properly interpret laboratory measurements of the microwave and millimeter-wave opacity of the gases which elute from liquid sulfuric acid, as well as to model their relation within the Venus atmosphere. Our results in Section III provide this information.

Recently, we have developed models for microwave and millimeter-wave emission from the Jovian planets in order to evaluate how adjustments to ammonia abundance profiles affect the predicted emission spectrum. We have also compared these results with observations from earth-based radio telescopes. (In the future, we should be able to take advantage of the availability of data from several millimeter-wave radio telescope arrays in order to develop localized ammonia abundance profiles over the entire disk of one or more Jovian planets.) In the next year of Grant NAGW-533, we propose to use the results of our laboratory analysis of the millimeter-wave absorption from gaseous NH_3 under simulated Jovian conditions to complete a formulation which accurately predicts the opacity from gaseous ammonia in a Jovian-type atmosphere over the entire 1 mm to 20 cm

wavelength range (frequencies from 1.5 to 300 GHz). Further discussion of this and other work related to the outer planets is presented in Section II. Likewise, we plan to continue to make laboratory measurements which will support our interpretive work of the 13 cm absorptivity profiles in the Venus atmosphere, which we are developing as part of the Pioneer-Venus Guest Investigator program, as well as interpreting microwave and millimeter-wave emission measurements of Venus. These data sets will be invaluable for characterizing the spatial and temporal variabilities of H_2SO_4 in the Venus atmosphere (see Section III). Finally, we will complete designs for laboratory instrumentation which will allow us to measure the microwave and millimeter-wave properties of liquids and solids under simulated planetary conditions.

II. OUTER PLANETS STUDIES

The basic configuration and technique for conducting the measurement of millimeter-wave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in Semiannual Status Report #11 for Grant NAGW-533. As stated in the report, measurements of the absorptivity of gaseous ammonia under simulated Jovian conditions have been completed at both Ka-Band (32-40 GHz) and W-Band (94 GHz). In the last part of the grant year, we have completed the analysis of all laboratory data from these experiments.

We have compared our experimental data to various theoretical formulations for ammonia absorptivity. The Van Vleck-Weisskopf (1945) lineshape is known to be accurate at low pressures (less than 1 atm). Zhevakin and Naumov (1963) derived

a different lineshape and found that their lineshape gave better results than the Van Vleck-Weisskopf theory when compared to experimental data for atmospheric water vapor absorption. This lineshape was also derived independently by Gross (1955) and is sometimes referred to as the kinetic lineshape. Ben-Reuven (1966) derived a more comprehensive lineshape which was found to be more accurate at higher pressures. Several modifications to the Ben-Reuven formalism (i.e. Morris and Parsons, 1970) have been made for applications in planetary science as more laboratory data under planetary conditions has become available. Recently, Spilker and Eshleman (1988) have derived new pressure and temperature dependences for the Ben-Reuven lineshape based on laboratory data under simulated Jovian conditions from high accuracy measurements at 9-18 GHz. Their formalism may be extrapolated to the frequencies and conditions of our experiments.

Figure 1 shows a graph of the four theoretical formulations which have been investigated for ammonia absorptivity from 1 to 1000 GHz (under the conditions of the Ka-Band experiment). These are labeled VVW for the Van Vleck-Weisskopf lineshape, BR-BG for the Berge and Gulkis (1976) formalism of the Ben-Reuven lineshape, BR-TS for the Spilker modified Ben-Reuven lineshape (private communication, 1988), and ZN-G for the Zhevakin-Naumov (1963) or Gross (1955) lineshape. The contribution from the rotational lines near 600 GHz has been included. All calculations employ center frequencies and line intensities given by Poynter and Kakar (1975). The two Ben-Reuven formalisms also include the modification by Morris and Parsons (1970).

Figure 2 shows the results of our Ka-Band experiment as compared to three of the theoretical lineshapes. The Spilker modified lineshape is not shown here, but

is essentially the same as the Gross (Zhevakin-Naumov) shape for this frequency range (see Figure 1). Since the data favors both the Gross lineshape and the Spilker formalism at these frequencies, it is not clear which is more appropriate. However, by making measurements at a higher frequency where the lineshapes are more distinct, it is possible to determine which of these lineshapes is more appropriate. Figure 3 shows the results of our 94 GHz experiment as compared to all four theoretical lineshapes. At this frequency it is clear that neither the Van-Vleck Weisskopf lineshape nor the Gross lineshape is adequate in describing the observed opacity.

Because the Spilker formalism is based on laboratory data which was taken at pressures greater than 1 atm, we cannot assume that it is valid at lower pressures. In fact, this model contains a singularity around 1 atm. Theory predicts that there should be a smooth transition from the Ben-Reuven formalism to the Van Vleck-Weisskopf formalism at some low pressure. However, it is not known at what pressure this transition occurs. Although absorptivity measurements were initially made at several pressures ranging from 1 to 2 atm, the sensitivity of our measurement apparatus was not great enough to detect ammonia absorption under experimental conditions at pressures near 1 atm. In fact, no reliable data at Ka-Band (32-40 GHz) was taken below 2 atm. However, at W-Band (94 GHz), one reliable data set at pressures near 1 atm was available. Careful analysis of the data taken at a pressure of 1 atm shows that although no significant absorption was detected, the error bars reflect an upper limit for the opacity due to ammonia. This upper limit is well below the Van Vleck-Weisskopf theory even at 1 atm. This analysis is significant in that it shows the transition to the Van-Vleck Weisskopf theory must occur at some pressure

pressure below 1 atm.

Another activity over the past grant year has been the development of a radiative transfer model of the Jovian atmosphere. This model will utilize our laboratory data and can also be used to evaluate the need for further laboratory measurements of other possible absorbers. The model is similar to that described in dePater and Massie (1985), where a simple spherical model has been used instead of an oblate spheroid. The model is written in PASCAL and can be run on any IBM compatible personal computer. With appropriate modifications, it can also be used to model the other Jovian planets. At the present time, we have a working model which we have used to generate simulated emission spectra. Over the next several months, we plan to further improve the model so that we may evaluate the effects of various microwave and millimeter-wave absorbers.

The temperature-pressure profile for our Jovian model is shown in Figure 4, where we have tried to match the profile given by dePater and Massie (1985) as closely as possible. The dePater and Massie profile is based on Voyager data from Lindal et al. (1981) with an adiabatic extrapolation to the deeper levels of the atmosphere. We have also matched their ammonia abundance profile in order to see how our model compares with theirs. The ammonia abundance profile is shown in Figure 5. A sample output from our model is shown in Figure 6. This model utilizes the Ben Reuven (Berge and Gulkis, 1976) formalism for ammonia absorption. When all of the assumptions of dePater and Massie are matched, we find that our spherical model differs from their oblate spheroid model by less than 5K (less than 3%) at all wavelengths from 1 mm to 10 cm (3-300 GHz). This is certainly within the error bars of the actual radio observations at these

wavelengths.

The next step will be to develop some kind of formulation for computing the absorption from ammonia at all temperatures, pressures, and frequencies based on all of the available laboratory data. Because laboratory data exists only at a few frequencies and at pressures above 1 atm, it is impossible to develop a single expression which will work under all conditions. The Spilker model provides a good estimate for the frequency dependence of the ammonia inversion spectrum under Jovian conditions at pressures around 2 atm. However, because of a singularity near 1 atm, it cannot be used in the model at all pressures. Instead of trying to use one single formulation for ammonia absorptivity which works at all temperatures, pressures, and frequencies, we will try a different approach. We will use the different theoretical and empirical formulations (i.e. those in Figure 1) over the entire frequency spectrum, but only at pressures and temperatures where they are known to be accurate. Since the abundance profile of ammonia contains abrupt changes at cloud layers, we may change the absorption spectrum at points in the model which will not cause severe discontinuities. For example, we may use either the Van Vleck-Weisskopf or the Ben-Reuven lineshape to describe ammonia absorption in the upper atmosphere above the main cloud layer (pressures below 0.7 atm.), where ammonia is depleted and the overall pressure is low. Since the contribution to the brightness temperature is negligible at these pressures, the assumed lineshape will not affect the calculated brightness temperature. The Berge and Gulkis formulation of the Ben-Reuven lineshape may be used from the main cloud layer to pressures above 1 atm. where the mixing ratio of ammonia levels off. The Spilker formalism may then be used at pressures above 1 atm., where it is known to be accurate.

We attempted to run our model using this approach. However, the Spilker formalism failed at higher frequencies for pressures greater than 1 atm. Further analysis of the Spilker formalism is needed before this lineshape can be used at higher frequencies and at pressures between 1 and 2 atm. We will continue to develop a model for ammonia absorption, based on laboratory results, which can be used in the emission model.

At the present time, the laboratory results can be used to give a lower limit for the abundance of ammonia on Jupiter. Since our laboratory results show that ammonia is less opaque than the Ben-Reuven lineshape predicts, the models of brightness temperature produced using the Ben-Reuven lineshape give a lower limit for the predicted brightness temperature. In other words, model output using the laboratory data will give higher brightness temperatures than those using the Berge and Gulkis (1976) Ben-Reuven formalism. DePater and Massie (1985) showed that more opacity was needed in order to account for the cooler brightness temperatures observed at millimeter-wavelengths. Thus, our laboratory results show that more ammonia must be present or other absorbers must exist in order to explain the observations.

Figure 7 shows several weighting functions for our Jovian model using the Berge and Gulkis model for ammonia absorption. At the peak of the ammonia inversion spectrum at 23 GHz, the main contribution to the brightness temperature comes from the layers just below the high-altitude crystalline ammonia cloud layer. At 40 GHz (Ka-Band), the main contribution centers around 1 atm. The weighting functions for 10 GHz and 94 GHz are almost identical. Since laboratory data has

of this figure shows that by using higher concentrations of H_2S , absorption could be measured with our laboratory configuration.

Larson et al. (1984) have developed a profile for the vertical distribution of hydrogen sulfide for Jupiter. This profile is shown with the ammonia profile in Figure 5. An ammonium hydrosulfide cloud (NH_4SH) is proposed around 2 atm, above which H_2S is depleted and below which a solar abundance of H_2S exists. The model was run using this profile along with our formulation for hydrogen sulfide absorption. The model spectra with and without hydrogen sulfide absorption are shown in Figure 9. The hydrogen sulfide spectrum should be slightly smoothed as in Bevard et al. (1983). However, this does give an upper limit on the effect of adding this H_2S profile to the model. This profile is also a lower limit on the amount of H_2S which may be present in the atmosphere of Jupiter. Several researchers (e.g. Briggs and Sackett, 1989) have suggested that as much as ten times the solar amount of H_2S may be present on the Jovian planets. A third model emission spectrum resulting from an H_2S profile similar to that in Figure 5, but with ten times the solar abundance below the NH_4SH cloud is also shown in Figure 9. Note that at the H_2S line centers, the calculated brightness temperature is not dependent on the H_2S mixing ratio, but is dependent on the level at which the ammonium hydrosulfide cloud is placed. This is similar to the effect at the center of the ammonia inversion spectrum at 23 GHz, where the brightness temperature is dependent on the level of the ammonia cloud layer as shown by the weighting function in Figure 7.

Over the next several months, we plan to further investigate the effects of different H_2S and NH_3 abundance and absorption profiles on the calculated

emission spectra. We will also investigate the possible effects of cloud condensates on the millimeter-wave emission. We will then evaluate the need for laboratory measurements of gaseous H_2S and other possible liquid absorbers.

III VENUS STUDIES

The laboratory measurement of the dissociation factor of gaseous H_2SO_4 in equilibrium with liquid sulfuric acid is desperately needed in order to allow proper interpretation of both future and existing work on the opacity of gaseous H_2SO_4 and to properly model its saturation abundance in the Venus atmosphere. Steffes (1985,1986), estimated that about 47% of the gaseous H_2SO_4 which vaporized from a liquid sulfuric acid reservoir, dissociated to form gaseous SO_3 and H_2O . Using this assumption, Steffes proceeded to calculate the mixing-ratio normalized opacity of gaseous H_2SO_4 , which is believed to be the major predominant microwave absorber at altitudes above 30 Km in the Venus atmosphere. The laboratory measurements used for calculating the normalized opacity were conducted at wavelength ranging from 1.3 to 22 cm. One wavelength of major interest is 13 cm. Its importance is due to the fact that measurements of localized absorptivity at that frequency have been provided by the Pioneer-Venus Orbiter Radio Occultation (ORO) experiment.

A. EXPERIMENTAL SETUP

The measurements are conducted using the apparatus shown in Figure 10. In this setup, a large vacuum chamber (of known volume), is constructed using Pyrex glass

as shown in Figure 11. The reason for choosing Pyrex is its ability to withstand high temperatures (600 K). The top of the glass chamber was sealed using a stainless steel plate. A high temperature O-ring was sandwiched between the glass and the plate to insure that leaks are minimal at that junction. A special purpose clamp is used to hold the plate to the glass lip. Stainless steel pipes and valves are used throughout the system in order to minimize acid-metal reaction at high temperatures. The pipes were submerged in sulfuric acid solution to dissolve surface impurities. The flask is also constructed of Pyrex glass. Another reason for choosing glass is the ability to monitor the status of the sulfuric acid vapor, and to make sure that no vapor condensation is occurring during pressure measurement.

The chamber and the flask had to be fitted inside a temperature controlled oven. The temperature inside the oven is monitored using a pre-calibrated thermocouple. The output of the thermocouple is connected to a digital voltmeter, the temperature is then inferred using voltage versus temperature data of the thermocouple. Two vacuum gauges are used in the experiment. Gauge P1 is used to monitor the status of the chamber, i.e. checks for leaks. The range on P1 is 0 - 20 Torr. Gauge P2 is connected to a digital pressure display. The gauge is able to measure pressures ranging from 0 to 800 Torr in 1 Torr increments, with accuracy of ± 1 Torr throughout its usable range. The latter gauge is used to measure total pressure in the chamber at the desired temperature. A nitrogen buffer is used in the setup so as not to allow any sulfuric acid vapor to come in contact with gauge P2. The buffer is filled with a predetermined amount of nitrogen. No buffer is necessary for gauge P1 because the valve is closed during pressure measurements. The experiment is conducted at six distinct temperature

points ranging from 490 to 600 K. Two different concentrations of liquid sulfuric acid are used, 99% and 95.9%. Acid samples were provided by the Du Pont Company. The flask is filled with a precisely known volume of liquid sulfuric acid at room temperature. The volume measurements are made using a 1-ml syringe with 0.01 ml gradations. For volumes less than 1 ml, repeatable accuracies of better than .01 ml have been obtained.

The apparatus is first heated to the desired temperature (usually taking about four hours). At that point, a vacuum is drawn. Using gauge P1 we are able to make sure that leaks are minimal, since any major leak can affect the total measured pressure, therefore altering the dissociation constant. Our system had a leak rate of approximately 1 Torr/hour. The system is allowed to reach thermal equilibrium, by continuously monitoring the temperature. The flask valve is then opened allowing the liquid H_2SO_4 to reach vapor pressure equilibrium with the evacuated chamber. Once equilibrium is reached, the valve is closed, and a check is made to verify that the remaining liquid is clear. The valve is usually opened for a period of ten minutes. The buffer is then opened and the chamber pressure is recorded. It is important to emphasize that throughout this process the temperature had to be within certain limits of the desired value, thus allowing the temperature variation factor to be minimal. In this case we were able to maintain oven temperature to within ± 5 K. The system is then allowed to cool overnight to its original room temperature. The remaining volume of liquid sulfuric acid is measured and the evaporated volume is inferred.

B. RELATION BETWEEN MEASURED QUANTITIES AND RESULTS

Let V_{Liquid} (ml) denote the volume of liquid H_2SO_4 and liquid H_2O which evaporates from the flask in Figure 10.

$$V_{\text{Liquid}} = V_{\text{H}_2\text{SO}_4} + V_{\text{H}_2\text{O}} \quad (1)$$

The percent concentration of the liquid sulfuric acid (by mass) is defined as follows : (i.e. PCM = .99 for 99% concentration by mass)

$$\text{PCM} = \frac{V_{\text{H}_2\text{SO}_4} \rho_{\text{H}_2\text{SO}_4}}{V_{\text{H}_2\text{SO}_4} \rho_{\text{H}_2\text{SO}_4} + V_{\text{H}_2\text{O}} \rho_{\text{H}_2\text{O}}} \quad (2)$$

where $\rho_{\text{H}_2\text{O}}$ = density of H_2O (1 gm/ml). From equation (1) and (2) one can solve for $V_{\text{H}_2\text{O}}$ and $V_{\text{H}_2\text{SO}_4}$:

$$V_{\text{H}_2\text{O}} = \frac{V_{\text{H}_2\text{SO}_4} \rho_{\text{H}_2\text{SO}_4} (1 - \text{PCM})}{\text{PCM}} \quad (3)$$

and

$$V_{\text{H}_2\text{SO}_4} = \frac{V_{\text{Liquid}} \cdot \text{PCM}}{\text{PCM} + \rho_{\text{H}_2\text{SO}_4} (1 - \text{PCM})} \quad (4)$$

In equations (3) and (4), the density of liquid H_2O have been assumed to be 1 gm/ml. Given the density of liquid sulfuric acid (gm/ml), it is possible to compute the number of H_2SO_4 molecules which are converted to vapor phase by the relation :

$$n_{\text{vapor}} = (V_{H_2SO_4} \cdot \rho_{H_2SO_4})/98 + (V_{H_2O} \cdot \rho_{H_2O})/16 \quad (5)$$

where n_{vapor} is the number of molecules (in moles) which vaporized during the experiment. The total number of molecules in the chamber (n_{total}) which resulted from the vaporized molecules can be determined assuming a dissociation factor D:

$$n_{\text{Total}} = \left[\frac{V_{H_2SO_4} \cdot \rho_{H_2SO_4}}{98} \right] (1+D) + \left[\frac{V_{H_2O} \cdot \rho_{H_2O}}{16} \right] \quad (6)$$

where D is the portion of H_2SO_4 molecules which dissociate to form SO_3 and H_2O . (The range of D is between 0 and 1). The pressure P (Atm) obtained from the evaporated liquid sulfuric acid can be directly measured during the experiment. P is related to n_{total} by the ideal gas equation :

$$P V_{\text{Chamber}} = n_{\text{Total}} \cdot R \cdot T \quad (7)$$

V_{chamber} (l) is the volume of the chamber occupied by the evaporated gas. R is the ideal gas constant (.08205 l.Atm/mole.K), and T is the temperature of the chamber in Kelvins (K). Combining the above equations, and solving for the dissociation

constant D yields :

$$D = \frac{784 \cdot \text{PCM} \cdot P \cdot V_{\text{chamber}} + V_{\text{H}_2\text{SO}_4} \cdot \rho_{\text{H}_2\text{SO}_4} \cdot R \cdot T \cdot (41 \text{ PCM} - 49)}{8 (\text{PCM}) V_{\text{H}_2\text{SO}_4} \cdot \rho_{\text{H}_2\text{SO}_4} \cdot R \cdot T} \quad (8)$$

Thus if the chamber volume, temperature of the system, density of sulfuric acid, pressure resulting from the evaporated liquid acid, and the amount of evaporated liquid are known, by using equation (8) one can compute the dissociation factor D.

Another important derivation is the calculation of the partial pressure of sulfuric acid ($P_{\text{H}_2\text{SO}_4}$ (Atm)). The calculation of the partial pressure, will enable us to go back and correct the measured normalized microwave absorptivity (Steffes, 1985) for gaseous H_2SO_4 in a CO_2 atmosphere at 13.4 and 3.6 cm. wavelength. The total pressure P can be written as :

$$P = P_{\text{H}_2\text{O}} + P_{\text{SO}_3} + P_{\text{H}_2\text{SO}_4} \quad (9)$$

Since $P_{\text{H}_2\text{O}} = P_{\text{SO}_3}$, (9) becomes :

$$P = P_{\text{H}_2\text{SO}_4} + 2 P_{\text{SO}_3} = P_{\text{H}_2\text{SO}_4} + 2 P_{\text{H}_2\text{O}} \quad (10)$$

The dissociation factor D can also be written as :

$$D = \frac{P_{\text{SO}_3}}{P_{\text{H}_2\text{SO}_4} + P_{\text{SO}_3}} \quad (11)$$

Combining (10) and (11) yield :

$$P_{\text{H}_2\text{SO}_4} = P \cdot \frac{1 - D}{1 + D} \quad (12)$$

Thus the partial pressure of sulfuric acid can be computed from the knowledge of the total pressure measured and the calculated dissociation factor D using (8). Note that when the dissociation factor is unity (10) and (12) yield :

$$P = 2 P_{\text{SO}_3} = 2 P_{\text{H}_2\text{O}} \quad (13)$$

Knowing the volume of the evaporated liquid and the pressure in the chamber the dissociation factor D can be calculated. Using D, the partial pressure of gaseous sulfuric acid can then be calculated using (12).

C. RESULTS

Figure 12 is a plot of the calculated partial pressure of gaseous sulfuric acid above liquid sulfuric acid as a function of temperature for a 99% (by weight) concentration. Illustrated points are from laboratory measurements. The solid

line is the partial pressure of sulfuric acid as calculated by Steffes (1985). Dashed line is a best-fit curve to the lab measurements. The temperature dependence of the vapor pressure of gaseous sulfuric acid is given by :

$$\ln p = 2.72 - 3952/T \quad (14)$$

where p is the sulfuric acid vapor pressure in atmospheres and T is the temperature in Kelvins. A similar result is obtained for a 95.9% concentration as shown in Figure 13. Its best-fit expression is given by (15):

$$\ln p = 2.89 - 4132/T \quad (15)$$

Figure 14 compares the best-fit results from these measurements with those obtained by Steffes (1985). Examination of our results indicate that the dissociation factor increases as the concentration is decreased. This is to be expected, since lower concentration means less sulfuric acid vapor pressure. Notice that the slope is constant for both concentrations, which suggests that partial pressure is proportional to concentration. Thus as concentration of sulfuric acid decreases so does the vapor pressure. Our results indicate that Steffes (1985) overestimated partial pressure at higher temperatures, and underestimated partial pressure at lower temperatures.

Although the above results are very critical in interpreting future and present work on the opacity of gaseous sulfuric acid, we need to further study the partial pressure of H_2SO_4 at lower temperatures. Thus, our results in this present form cannot currently be extrapolated to lower temperatures. The results

from lower temperatures studies will allow us to develop a single expression for partial pressure of gaseous H_2SO_4 over a larger temperature range. In the next grant year, we will go back to the results obtained by Steffes (1985) and correct the normalized microwave absorptivity of gaseous sulfuric acid at 13.4 and 3.6 cm, in order obtain new sulfuric acid mixing ratio profiles for the Venus atmosphere. Our results can also be used to infer the saturated sulfuric acid abundance by using published data on the pressure-temperature dependence of the Venus atmosphere (Seiff et.al, 1980) along with our results. Since total pressure is known and saturated partial pressure can be calculated using our formulas, mixing ratio is then easily calculated.

IV. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

Over this past grant year, a paper was published in Icarus, describing results and applications of the laboratory measurements of the millimeter-wave opacity of ammonia between 7.5 and 9.38 mm, described in previous reports (Joiner et al., 1989). In addition, we completed a paper describing observations and interpretive studies of the 1.3 to 3.6 cm Venus emission spectrum (Steffes et al., 1989), which was also accepted for publication in Icarus. We also submitted updated summaries of our most recent laboratory measurements for inclusion in the twenty-third issue of the Newsletter of Laboratory Spectroscopy for Planetary Science.

We will attend the 21st Annual Meeting of the Division for Planetary Sciences of the American Astronomical Society the week of October 30 through November 4, 1989. We will present 4 papers (abstracts attached--see Appendix A.) Three of these papers will address laboratory measurements and interpretive studies of

the Venus atmosphere, and one will present results of millimeter-wave studies of the Jovian atmosphere.

In addition to the radio astronomical observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked with Dr. Michael A. Janssen of JPL regarding models for the Venus atmosphere and the outer planets, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of H_2SO_4 . We have also worked with Dr. Arvydas J. Kliore of JPL on the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies as part of our involvement in the Pioneer-Venus Guest Investigator Program. More informal contacts have been maintained with groups at the California Institute of Technology (Drs. Duane O. Muhleman, Kathryn Pierce, and Arie Grossman, regarding interpretation of radio astronomical measurements of Venus and the outer planets). We have also worked closely with the Stanford Center for Radar Astronomy (Drs. V.R. Eshleman, G.L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using out laboratory measurements of atmospheric gases in the interpretation of radio astronomical observation of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets' atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (Goddard SFC).

During the Voyager-Neptune encounter, we supported the Voyager Radio Science Team

by providing calculations of the expected atmospheric absorption by gaseous H_2S and NH_3 in the Neptune atmosphere, at the Voyager S-Band and X-Band downlink frequencies. These calculations were used both for pre-encounter operational planning and post-encounter interpretive studies.

Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA (both as a "by-mail" reviewer and as a member of the February 1989 review panel) and as a reviewer of manuscripts submitted to Icarus, the Journal of Geophysical Research, Physics Reports, and Annual Review of Astronomy and Astrophysics. We have also continued to serve the planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the mission planning for radio and radar systems aboard the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Dr. Steffes also participated as a member of the International Jupiter Watch (IJW) Laboratory/Theory Discipline Team. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for Dr. Steffes' attendance at PAMOWG meetings, as well as the November AAS/DPS meeting, has been provided by Georgia Tech in support of Planetary Atmospheres Research. Also, in support of Planetary Atmospheres Research, Georgia Tech has provided a Hewlett-Packard Vectra QS-16 Computing System.

As in the past, we have maintained contact with members of the Georgia

congressional delegation, keeping them aware of our work and aware of our continued support for the solar system exploration program. We were especially pleased with the support received from Senator Wyche Fowler for the CRAF/Cassini "new start," after briefing his staff on this issue. (See Appendix B)

V. CONCLUSION

Over the past grant year, we have continued our work with laboratory measurements and interpretation of the millimeter-wave properties of atmospheric gases contained in the outer planets. The results of our studies have been significant in that they indicate that the large opacities predicted by a number of workers at these wavelengths are indeed incorrect and that a form of the modified Ben-Reuven formalism for computing the millimeter-wave opacity from ammonia is correct. In the future, we plan to continue development of our models for Jovian millimeter-wave emission. We plan to further investigate the effects of the H_2S and NH_3 on the calculated emission spectra. We will also evaluate the need for laboratory measurements of gaseous H_2S and other possible liquid absorbers.

Our Venus studies over the past grant year have made it possible for us to measure the dissociation of gaseous H_2SO_4 into SO_3 and H_2O , which will aid in modeling of the Venus atmosphere, and will make it possible to correct previous laboratory measurements of the microwave opacity of gaseous H_2SO_4 . In the next grant year we intend to develop corrected expressions for the microwave opacity from gaseous H_2SO_4 which will allow a more accurate interpretation of the 13 cm absorptivity profiles provided by Pioneer-Venus radio occultation studies. We will likewise complete designs and begin laboratory measurement of the millimeter-wave properties of liquid H_2SO_4 .

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VII. Key Figures

Figure 1: Theoretically calculated absorption from NH_3 under Jovian conditions using the Van Vleck-Weisskopf, Ben Reuven (Berge and Gulkis, and Spilker and Eshleman formalisms) and Zhevakin and Naumov or Gross lineshapes.

$T=210\text{K}$ $P=2\text{atm}$ $\text{XNH3}=1.8\text{e-}2$ $\text{XH2}=.9$ $\text{XHe}=.1$

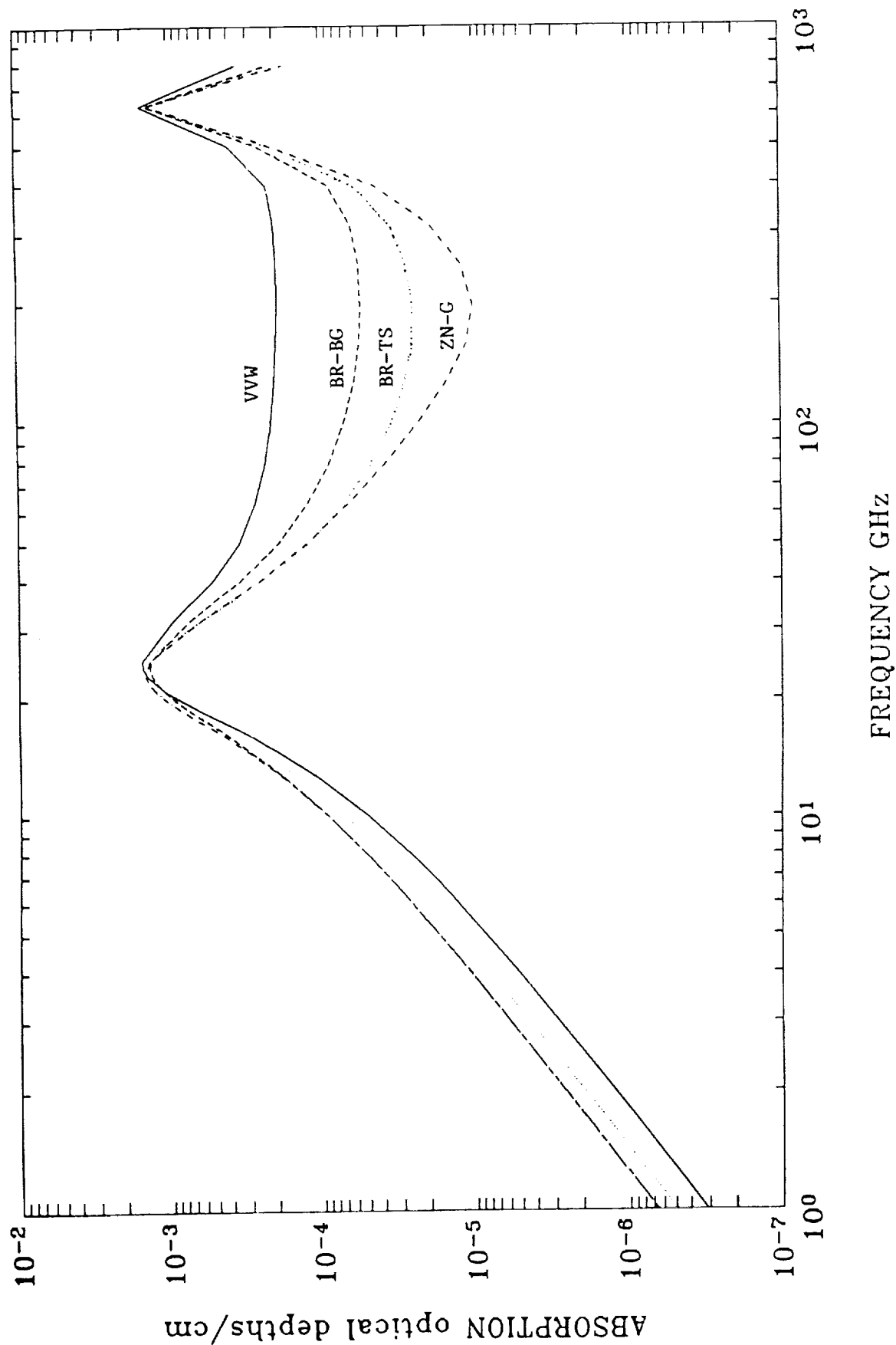


Figure 2:

Absorption of NH_3 in a
 88.34% H_2 , 9.81% He , 1.85% NH_3
 mixture (Mixing ratio: 0.0185 ± 0.0005)
 Pressure: 2 atm. Temp: 203K

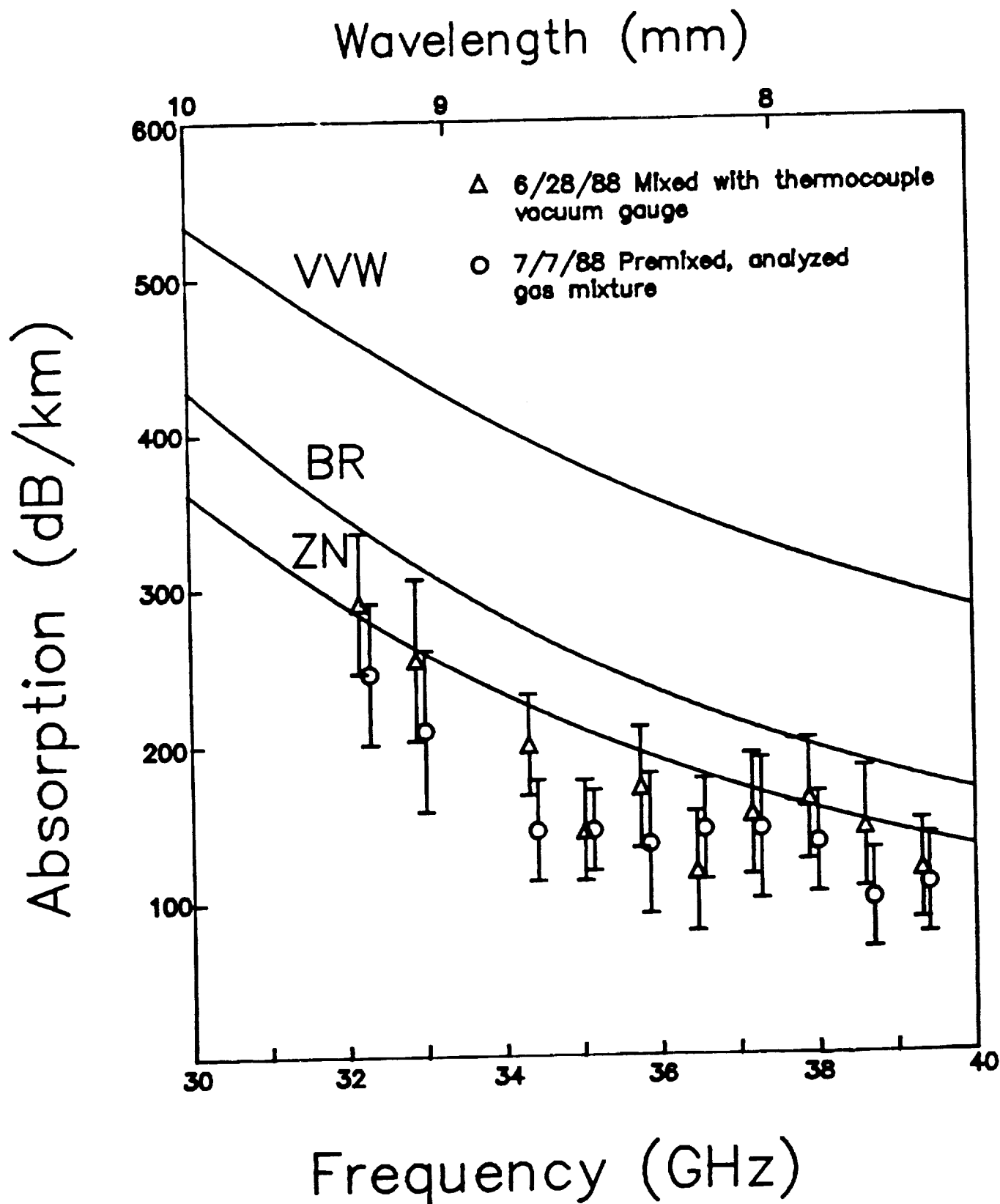


Figure 3: Measured ammonia (NH_3) absorption under Jovian conditions as compared to theoretically computed absorption.

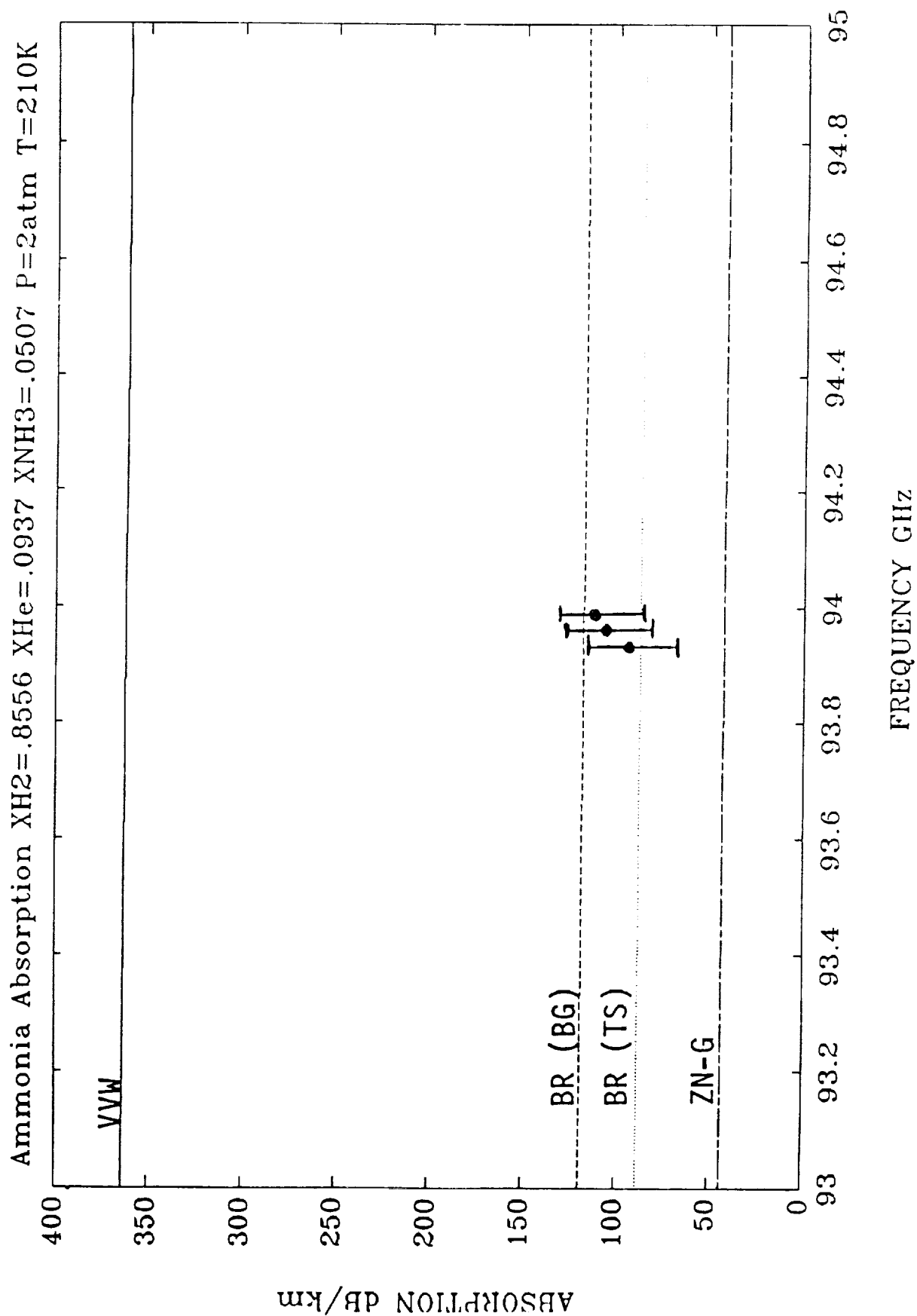


Figure 4: Jovian Temperature Pressure Profile (based on dePater & Massie)

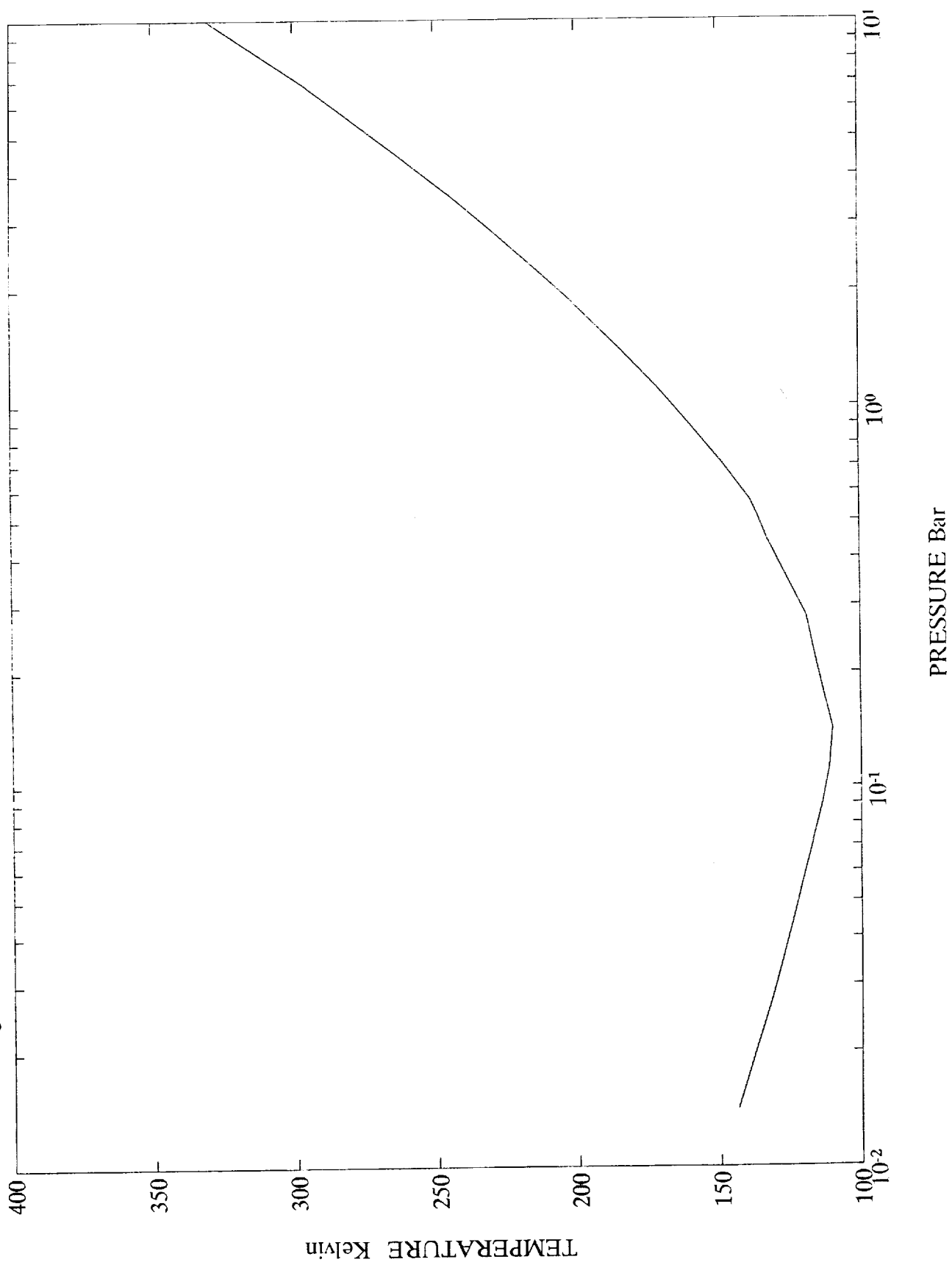


Figure 5: NH_3 and H_2S Mixing Ratios in Jovian Model

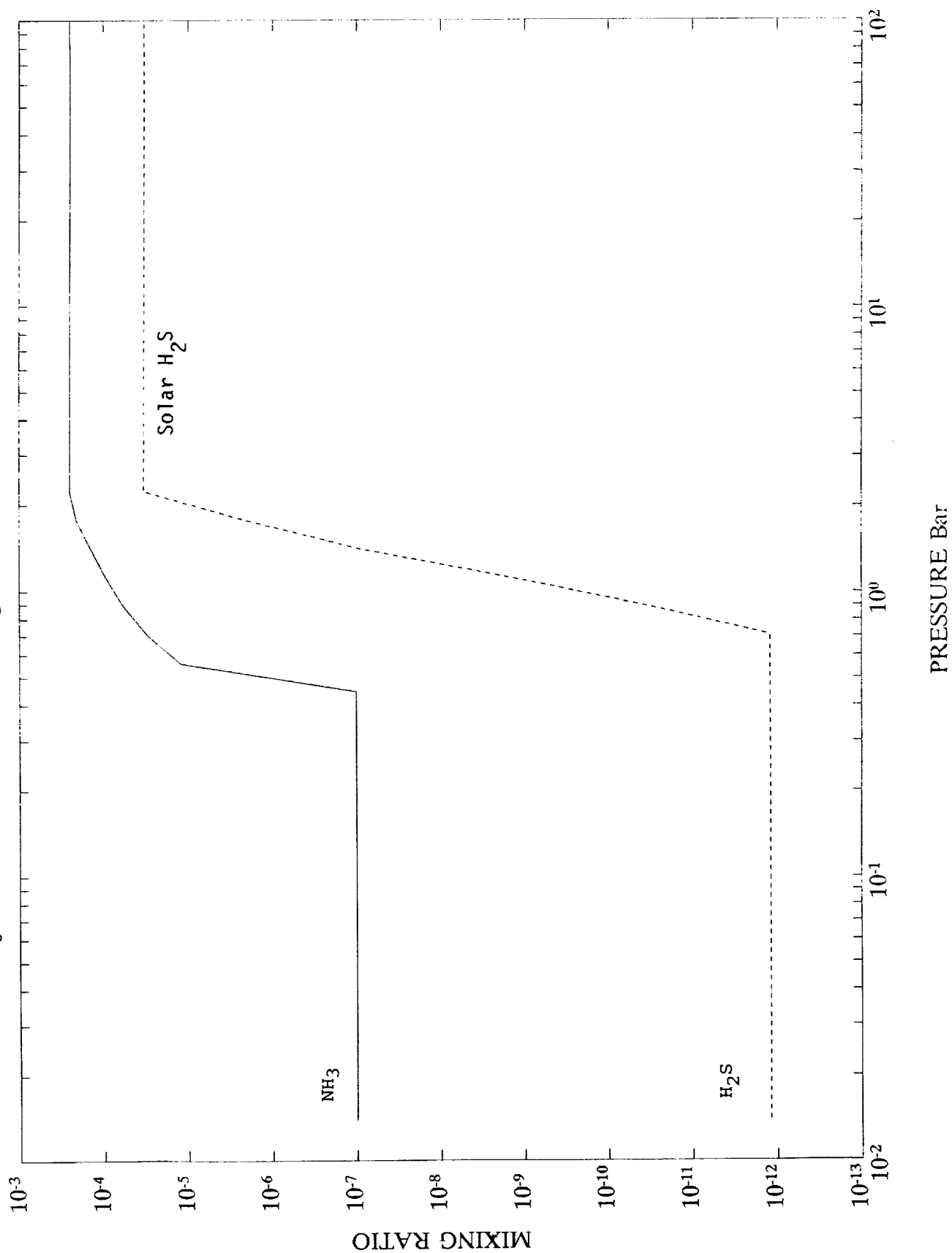
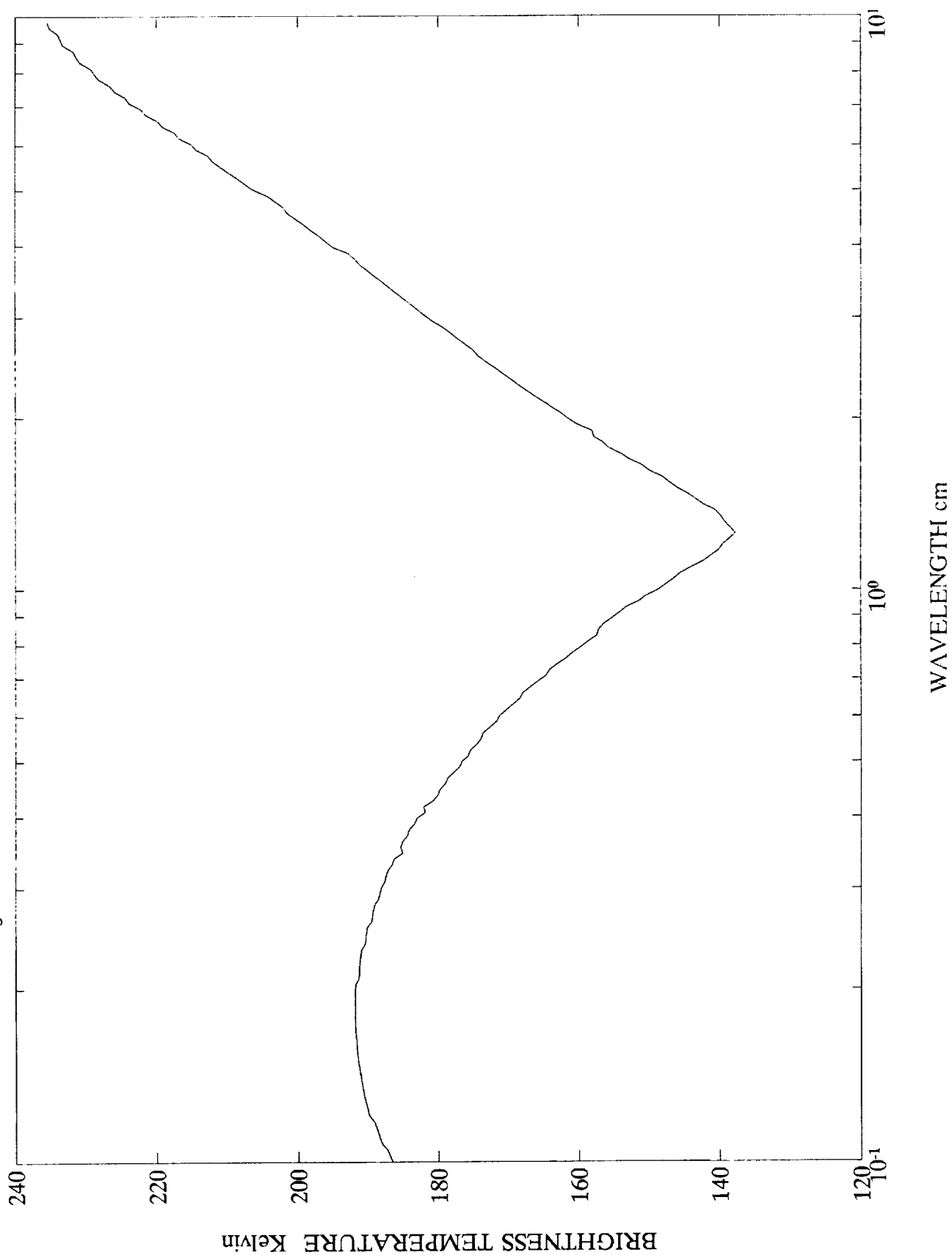


Figure 6: Jovian Model based on dePater & Massie 4BR NH_3 abundance as in Figure 5



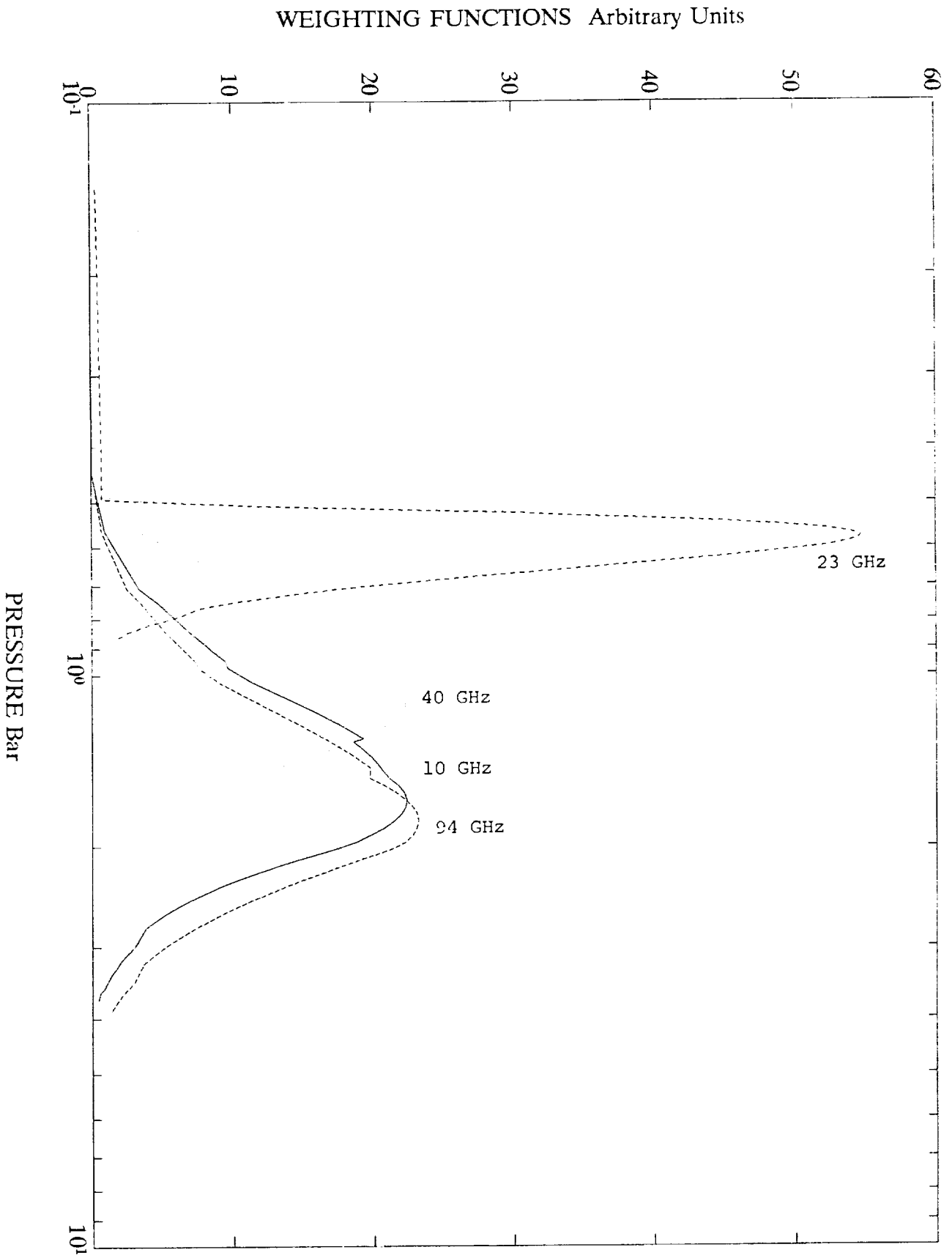


Figure 7: Weighting Functions for Jovian Model

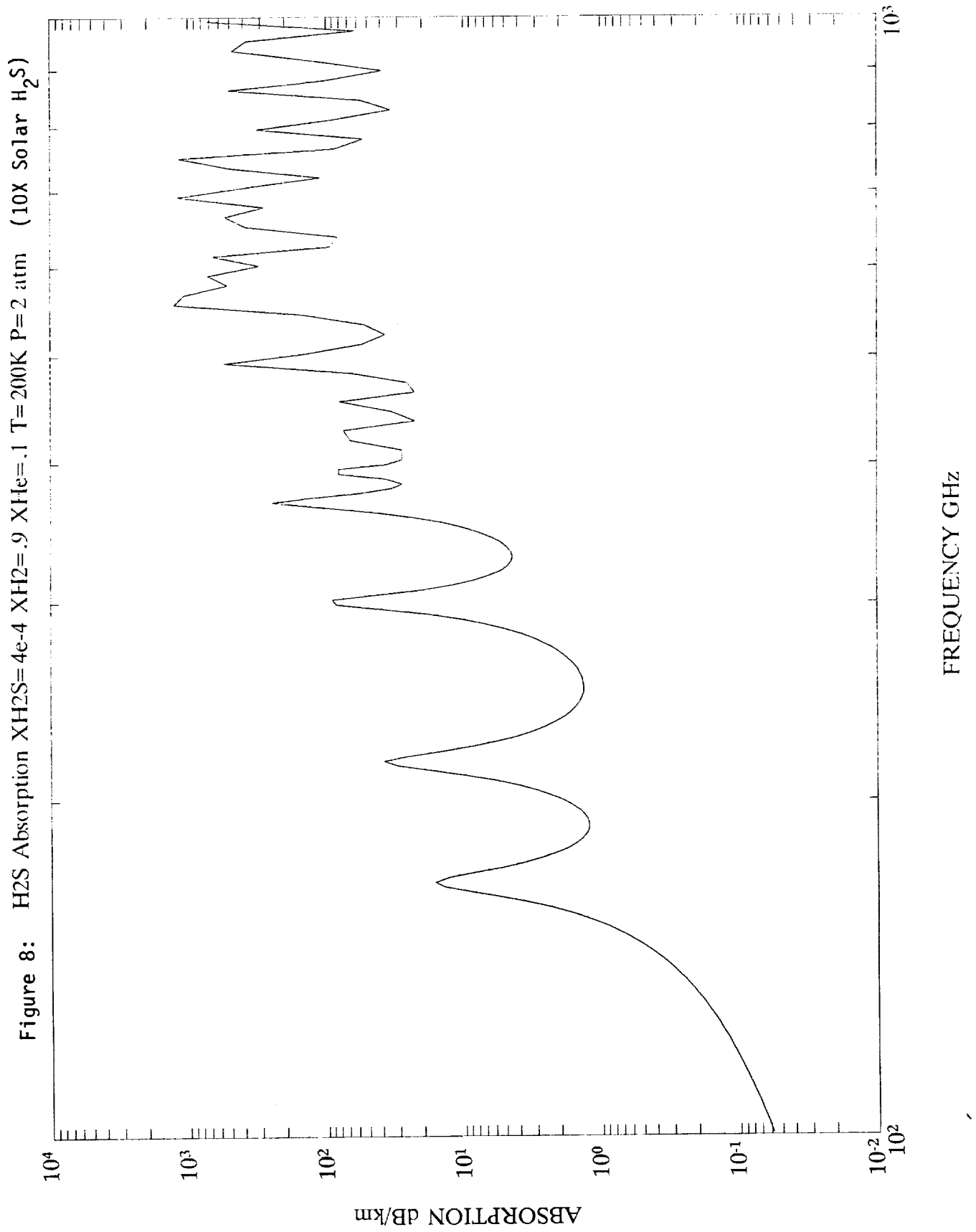
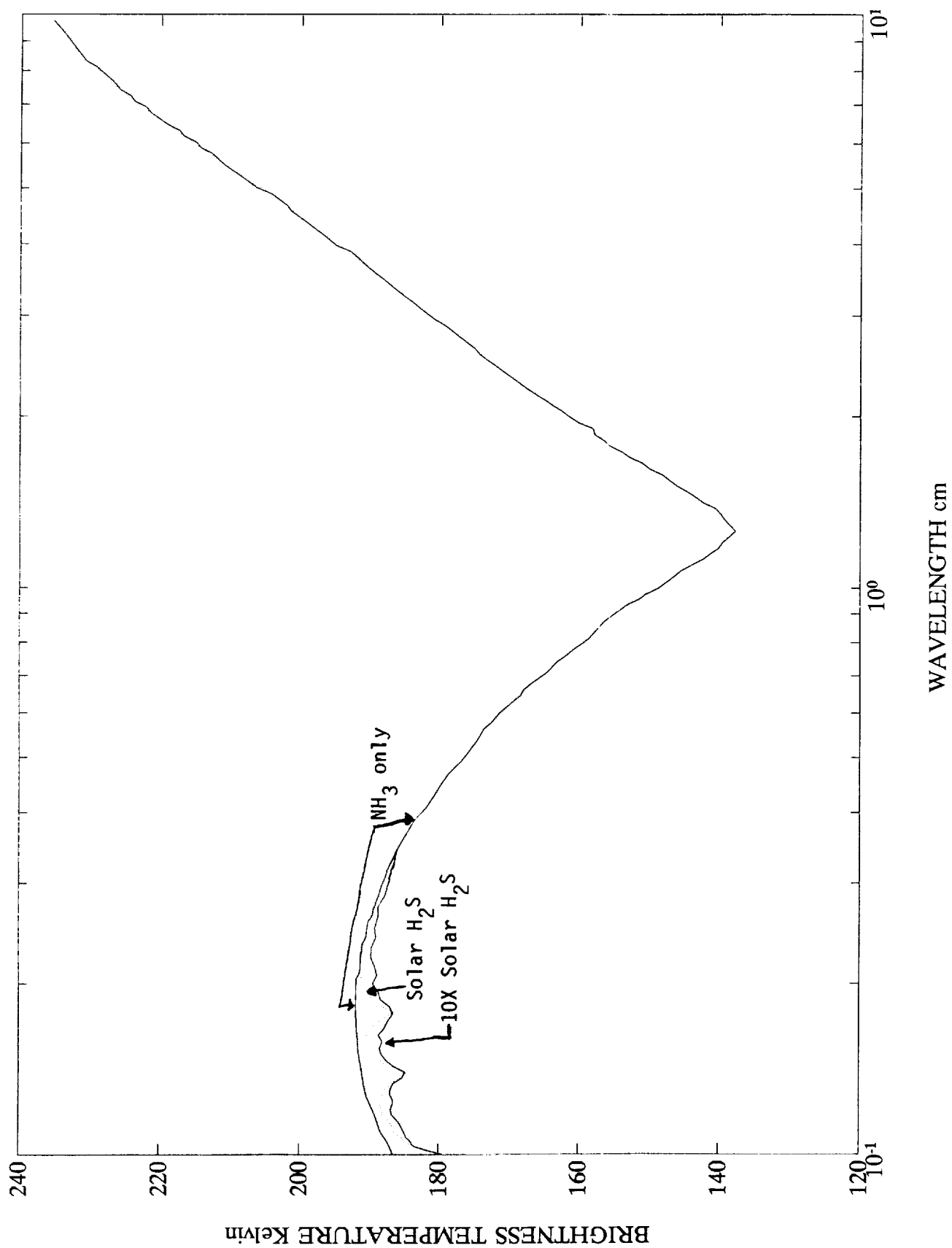


Figure 9: Jovian model with 1X and 10X Solar H₂S and no H₂S ammonia abundance as shown in Figure 5



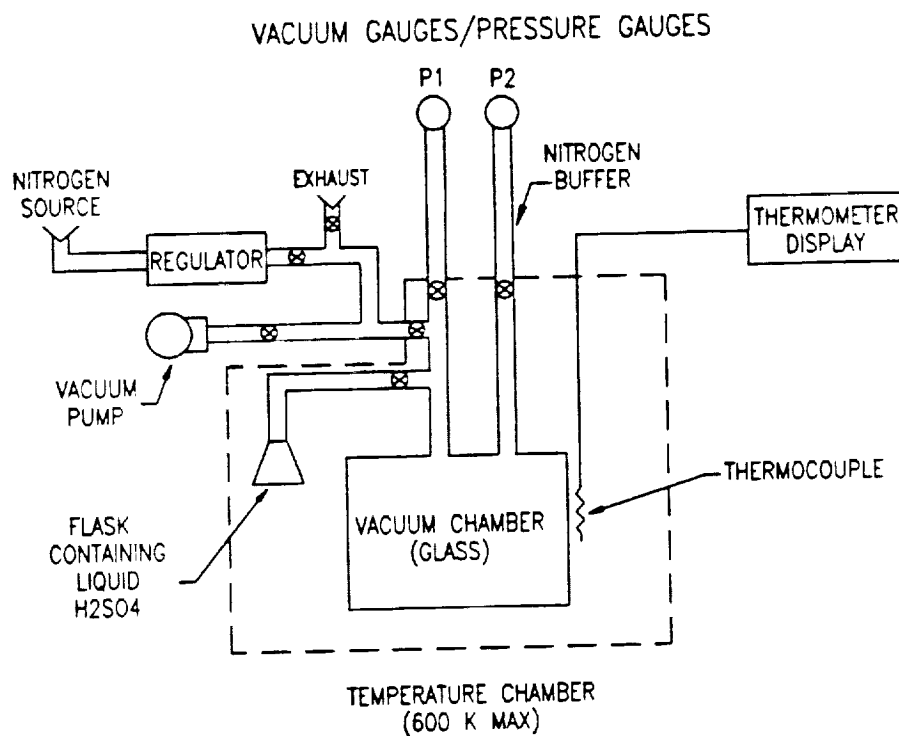


Figure # 10 : Laboratory apparatus used to measure the dissociation factor D , for H_2SO_4 above liquid sulfuric acid .

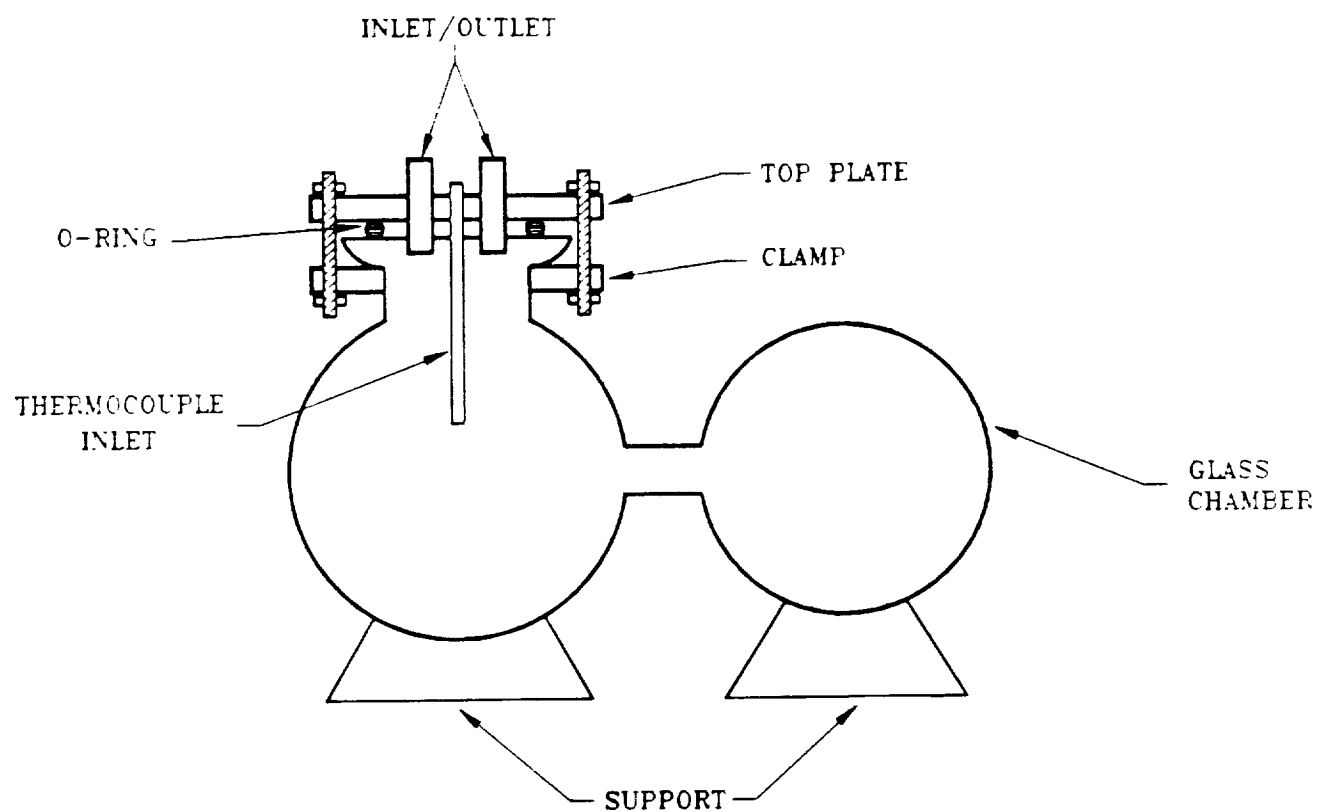


Figure # 11 : Cross section view of the vacuum chamber used in our apparatus .

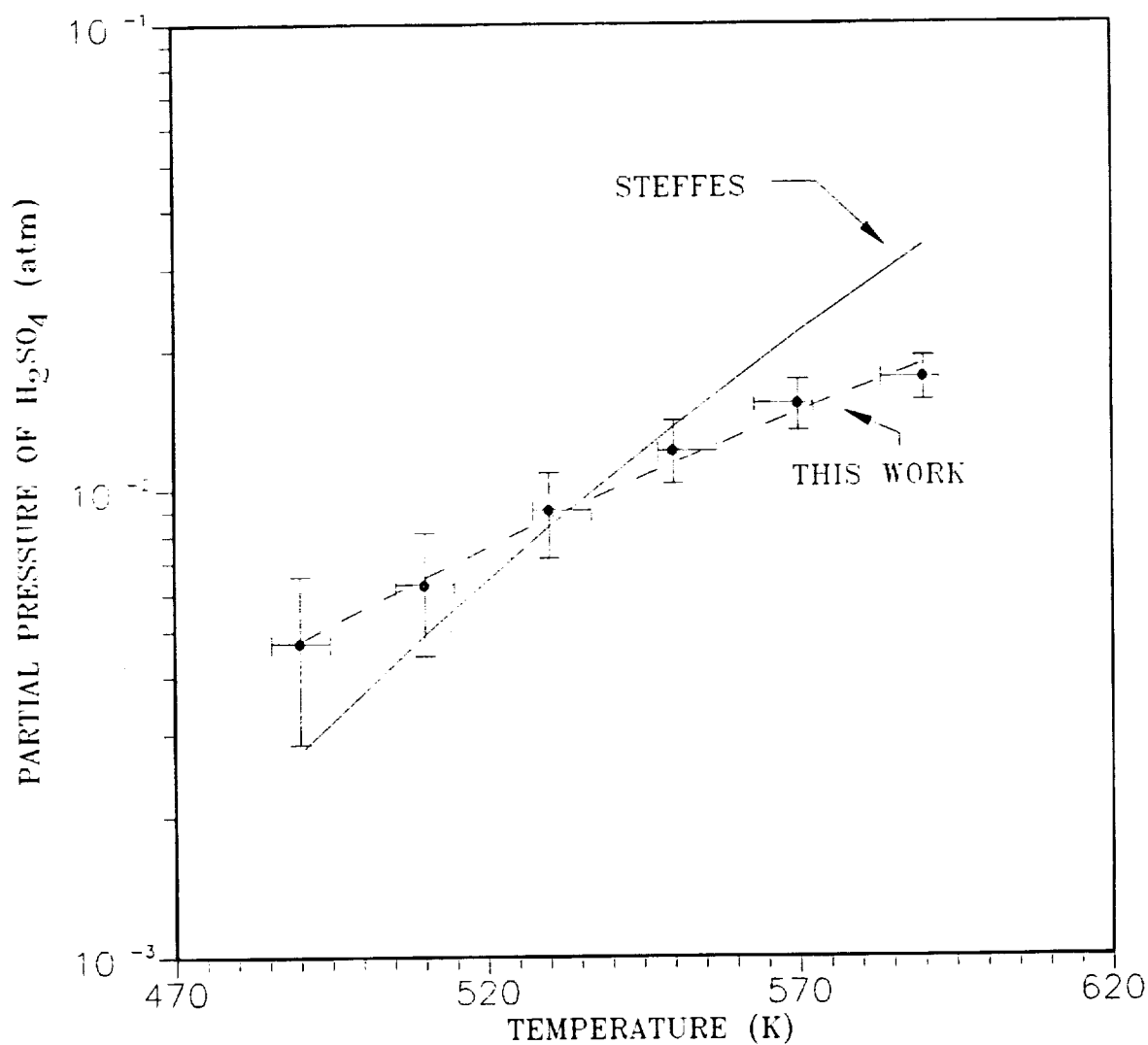


Figure # 12 : Vapor pressure of gaseous H_2SO_4 (99%) above liquid sulfuric acid as a function of temperature. The illustrated points are from laboratory measurements. Vapor pressure expression (solid line) from Steffes (1985) is compared with a best-fit expression for our measurements (dotted line). Error bars for pressure and temperature are shown.

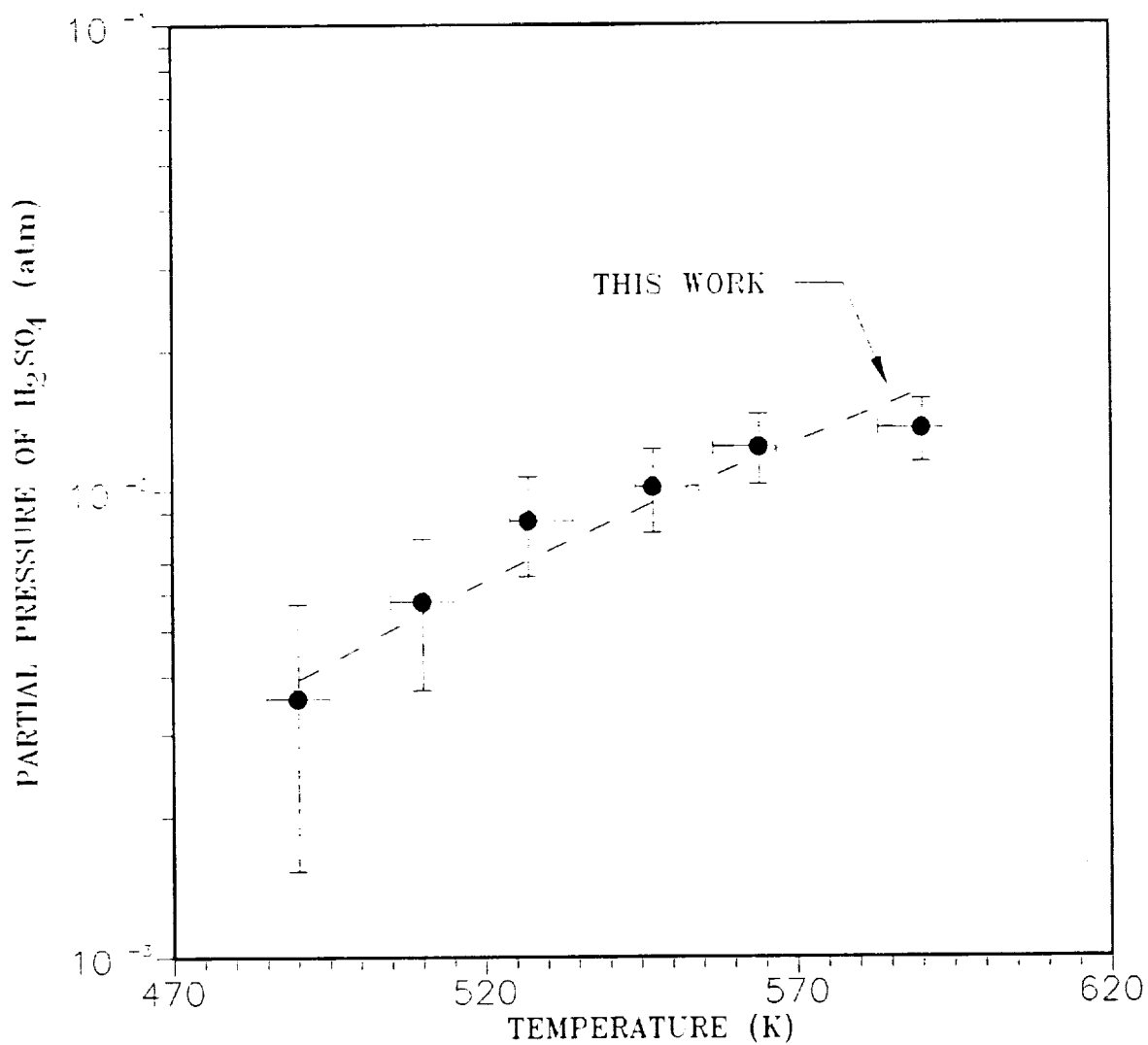


Figure # 13 : Vapor pressure of gaseous H_2SO_4 (95.9%) above liquid sulfuric acid as a function of temperature. Illustrated points are from laboratory measurements. Dotted line is a best-fit expression for our measurements. Error bars for pressure and temperature are shown.

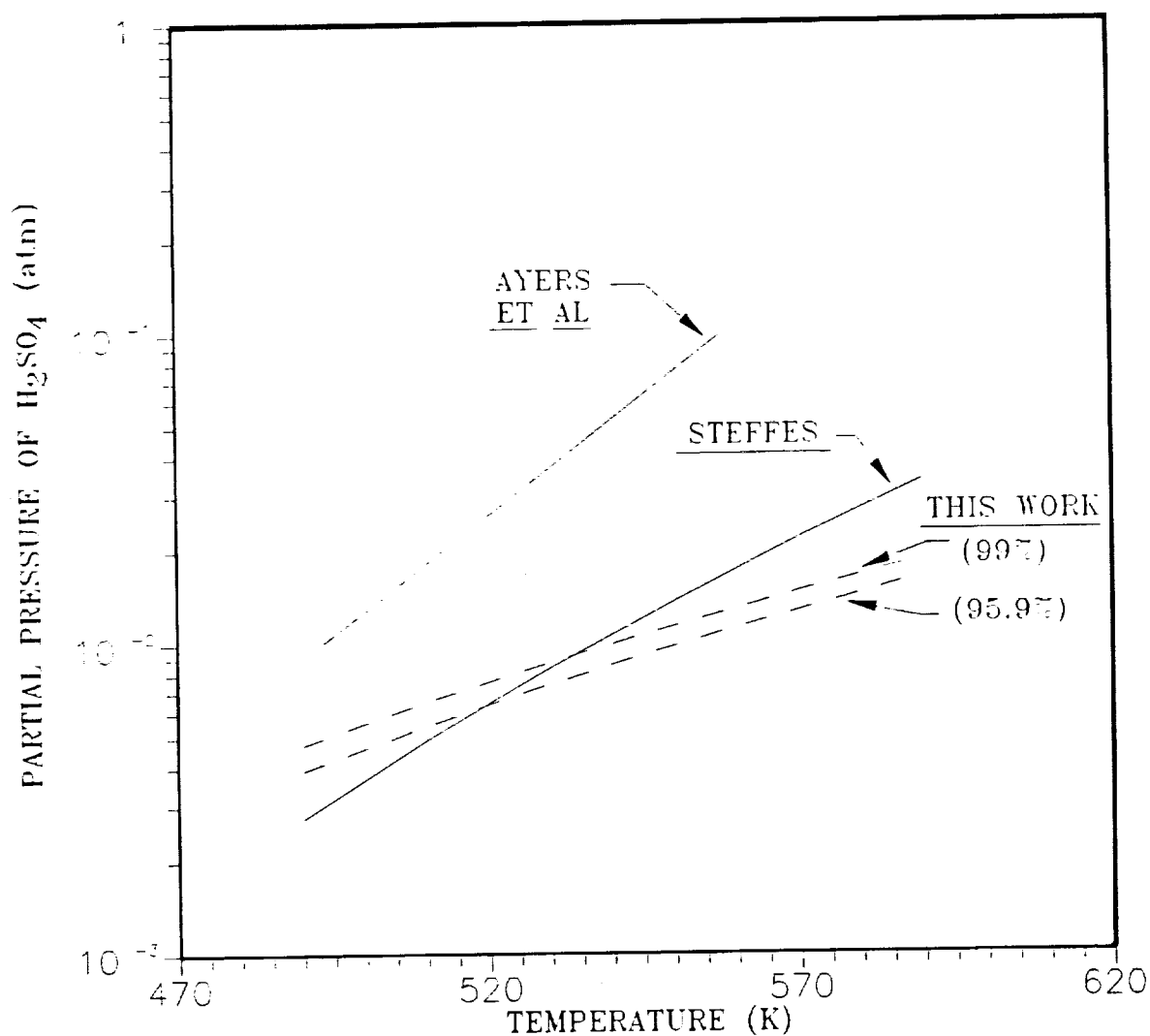


Figure # 14 : Best-fit expression (dotted line) for our measured partial pressure of H_2SO_4 as a function of temperature in comparison with vapor pressure expressions from Ayers et. al. (1980) and Steffes (1985).

Laboratory measurements of the dissociation factor of gaseous sulfuric acid (H_2SO_4)

A.K. Fahd, P.G. Steffes (Georgia Institute of Technology)

A good understanding of the dissociation of gaseous H_2SO_4 into SO_3 and H_2O is desperately needed in order to properly interpret laboratory measurements of the opacity of H_2SO_4 . The dissociation factor is also valuable in order to accurately model the saturation abundance of gaseous H_2SO_4 , SO_3 , and H_2O in the atmosphere of Venus.

Laboratory measurements of the dissociation factor of sulfuric acid vapor are described. The experiment is being conducted at temperatures ranging from 480 to 580 K. Two concentrations of sulfuric acid have been used, (98.7%, 99.0%). The partial pressures of water, sulfuric acid, and sulfur trioxide can be inferred from the measurements. A comparison between our results and those calculated by Gmitro and Vermulen (1964, *Amer. Inst. Chem. Eng.* 10, 740-746) will be presented.

*This work was supported by the Planetary Atmospheres Program of the National Aeronautics and Space Administration under grant NAGW-533.

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Potential Variability of the Abundance and Distribution of Gaseous Sulfuric Acid Vapor Below the Main Cloud Deck in the Venus Atmosphere.

J.M. Jenkins, P.G. Steffes (Georgia Institute of Technology)

Studies of recent measurements of the 1.35 to 3.6 μ m emission from Venus have suggested that long term temporal and/or spatial variations in the abundance of gaseous sulfuric acid vapor (H_2SO_4) may occur immediately below the main cloud layer (48 km and below). To investigate these issues, we have derived 13 cm absorptivity profiles from Pioneer-Venus Orbiter radio occultation data obtained in 1986 and 1987. Data from selected orbits have been analyzed which span a range of latitudes from 11°N to 88°N, solar zenith angles from 66° to 160°, and probe altitudes as deep as 40 km. In addition, upper limits on the abundance profiles of gaseous sulfuric acid vapor have been inferred from the absorptivity profiles. Furthermore, we have begun characterizing the uncertainties inherent in the data inversion process. When carried to its full potential, this radio occultation data will provide insight into possible variations in the abundance and distribution of gaseous H_2SO_4 below the main cloud layer in the Venus atmosphere.

*This work was supported by NASA Grant NAG 2-515. This material is also based on work supported under a National Science Foundation Graduate Fellowship.

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Models of the Millimeter-Wave Emission of the Jovian Atmosphere Utilizing Laboratory Measurements of Gaseous Ammonia (NH₃)

J. Joiner, P.G. Steffes (Georgia Institute of Technology)

Radiative transfer models of the Jovian atmosphere using the modified Ben-Reuven lineshape for the absorption of gaseous ammonia (NH₃) appear to deviate from observations at millimeter-wavelengths. Laboratory measurements at both 32-40 GHz (7.5 - 9.3 mm) and 94 GHz (3.2 mm) have been completed and confirm that some modified form of the Ben-Reuven lineshape does accurately describe the observed opacity of gaseous ammonia under simulated Jovian conditions. Models of the absorption spectrum of ammonia are being derived from laboratory measurements at both microwave and millimeter-wavelengths. Radiative transfer models are being developed which utilize these laboratory results. The models can also be used to evaluate other potential sources of millimeter-wave opacity such as cloud condensates and gaseous hydrogen sulfide (H₂S).

*This work was supported by the Planetary Atmospheres Program of the National Aeronautics and Space Administration under Grant NAGW-533.

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ABSTRACT FORM

Evidence for Temporal Variations in S₀₂ Abundance
in the Sub-Cloud Region of the Venus Atmosphere

P.G. Steffes (Georgia Institute of Technology)

Analysis of 1987 measurements of the microwave emission of Venus from 1.3 to 3.6 cm (Steffes, et al., 1989, Icarus, in press) places limits on the abundance of S₀₂ below the cloud layers (altitudes below 48 km). The S₀₂ abundance inferred from these measurements is a factor of 3-4 below the abundances measured in 1978 by the Pioneer Venus Sounder Probe (Oyama et al., 1980, JGR 85, 7891) and by the Venera 12 lander (Gel'man et al., 1979, Cosmic Research 17, 585). However, our result is in agreement with results from the Vega 1 and 2 Probes obtained in 1985 (Bertaux et al., 1987, Cosmic Research 25, 691). This strongly suggests a decrease in the subcloud abundance of S₀₂ in the period from 1978 to 1987, which parallels the observed decrease in S₀₂ at the cloud tops by Esposito et al., (1988, JGR 93, 5267) and by Na et al., (1988, B.A.A.S. 20, 832).

*This work was supported by the Planetary Atmospheres Program of the National Aeronautics and Space Administration under Grant NAGW-533.

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WYCHE FOWLER, JR.
GEORGIA

United States Senate

WASHINGTON, D.C. 20510

July 20, 1989

Mr. Paul G. Steffes
Associate Professor
Georgia Institute of Technology
School of Electrical Engineering
Atlanta, Georgia 30332

Dear Mr. Steffes:

Thank you for letting me know of your support for a strong American space program and for the CRAF/Cassini missions.

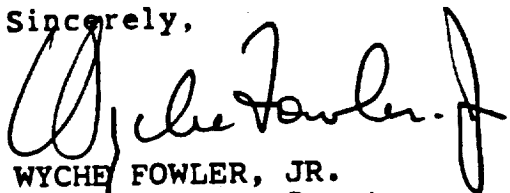
I share your interest in this field and have been a strong supporter of maintaining a vigorous space policy throughout my service in Congress. As a world leader in this area, I strongly believe we should strive to expand our knowledge of the solar system and exploit the opportunities such knowledge provides. One of the primary national goals should be to preserve America's leadership in space sciences and technology and their applications for peaceful purposes.

In addition to expanding our role in space, the national space program continues to develop technologies and uncover information that serve our immediate needs on Earth. Our activities in space have produced and continue to yield significant information and advances in several diverse fields, including communications, meteorology, and the global environment. Ongoing and future space projects have provided or will provide important information about the atmosphere, ozone depletion, the destruction of our tropical forests, and a host of other vital issues.

I am happy to report the Bush Administration has requested a \$13.3 billion dollar budget for NASA for fiscal year 1990. This is a 22% increase over the \$10.9 billion NASA received this year and will allow NASA to pursue its several projects on schedule. As the Senate Appropriations Committee examines the Administration's budget proposal and makes a final decision on the 1990 budget, you can count on my support for a strong American space program.

I appreciate your interest in this important subject.

Sincerely,



WYCHE FOWLER, JR.
United States Senator